



RECEIVED

JAN 13 2004

TC 1700

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
MASARU YOSHITAKE, ET AL. : EXAMINER: WILLS, MONIQUE
SERIAL NO: 09/889,776 :
FILED: JULY 23, 2001 : GROUP ART UNIT: 1746
FOR: POLYMER ELECTROLYTE FUEL CELL

DECLARATION

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes Masayuki KUWANO who deposes and says:

That my name is Masayuki KUWANO;

That my address is 5-24-310, 3-chome, Nakacho, Musashino-shi,
Tokyo, Japan;

That I know well both the English and Japanese languages;

That the attached English language translation is true and
correct translation of Japanese Patent Application No. 11-013437 filed on
January 21, 1999 to the best of my knowledge and belief;

I hereby declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

FURTHER DEPONENT SAITH NOT.

December 28, 2003
Date

Masayuki Kuwano
Masayuki KUWANO

Date of Application: January 21, 1999

Application Number: Patent Application JP11/013437

Applicant: Asahi Glass Company, Limited

【Type of Document】 PETITION FOR PATENT APPLICATION
【Reference No.】 980838
【Filing Date】 January 21, 1999
【Addressee】 Commissioner, Patent Office
【International Patent Classification】 H01M 8/04
H01M 8/10

【Inventor(s)】

【Name】 Masaru Yoshitake
【Address or Residence】 Asahi Glass Company, Limited
1150, Hazawa-cho, Kanagawa-ku,
Yokohama-shi, Kanagawa
【Name】 Eiji Yanagisawa
【Address or Residence】 Asahi Glass Company, Limited
1150, Hazawa-cho, Kanagawa-ku,
Yokohama-shi, Kanagawa
【Name】 Eiji Endoh
【Address or Residence】 Asahi Glass Company, Limited
1150, Hazawa-cho, Kanagawa-ku,
Yokohama-shi, Kanagawa
【Name】 Yasuhiro Kunisa
【Address or Residence】 Asahi Glass Company, Limited
1150, Hazawa-cho, Kanagawa-ku,
Yokohama-shi, Kanagawa

【Patent Applicant(s)】

【Identification No.】 000000044
【Name or Company Name】 Asahi Glass Company, Limited

【Representative(s)】

【Identification No.】 100090918
【Patent Attorney】
【Name or Company Name】 Kenji Senmyo

【Telephone Number】 03-3218-5647

【Identification of Fees】

【Prepayment account No.】 009830

【Amount of Payment】 21,000 yen

【List of Submitted Items】

【Name of item】	Specification	1
----------------	---------------	---

【Name of item】	Drawings	1
----------------	----------	---

【Name of item】	Abstract	1
----------------	----------	---

【Burden of Proof】 Yes

【TYPE OF DOCUMENT】 SPECIFICATION

【TITLE OF THE INVENTION】

POLYMER ELECTROLYTE FUEL CELL

【SCOPE OF THE CLAIM(S)】

5 【Claim 1】

A polymer electrolyte fuel cell comprising a plurality of membrane electrode assemblies laminated via separators, each assembly comprising a membrane-form polymer electrolyte and a pair of a fuel electrode and an
10 air electrode facing each other via the electrolyte, wherein the separator has a fuel gas channel for supplying a fuel gas to the fuel electrode, an oxidant channel for supplying an oxidant to the air electrode and a fluid channel for removing a heat generated by a
15 reaction out of the cell system, and the separator is made of a metal/non-metal composite material which has faces made of non-metal which are in contact with the membrane electrode assemblies and side walls of the fluid channel which are made of metal.

20 【Claim 2】

The polymer electrolyte fuel cell according to Claim 1, wherein the separator comprises a layer made of non-metal having the fuel gas channel on its surface, a layer made of metal having the fluid channel internally and a
25 layer made of non-metal having the oxidant channel on its surface, which are laminated, and the fuel gas channel

and the oxidant channel are disposed on the surface of the separator.

【Claim 3】

The polymer electrolyte fuel cell according to Claim
5 1 or 2, wherein the metal is a member selected from the group consisting of a metal containing aluminum in an amount of at least 80 wt%, a metal containing titanium in an amount of at least 80 wt% and stainless steel, and the non-metal comprises carbon material as the main component.

10 【Claim 4】

The polymer electrolyte fuel cell according to Claim 1, 2 or 3, wherein a coating film containing ceramics and having a resistivity of at most $3 \times 10^{-4} \Omega \cdot \text{cm}$, is formed on the surface of the side walls of the fluid channel.

15 【Claim 5】

The polymer electrolyte fuel cell according to Claim 1, 2, 3 or 4, wherein a layer containing ceramics and having a resistivity of at most $3 \times 10^{-4} \Omega \cdot \text{cm}$, is disposed at the interface between the metal and the non-metal.

20 【DETAILED DESCRIPTION OF THE INVENTION】

【Technical Field to which the Invention Belongs】

The present invention relates to a polymer electrolyte fuel cell.

【Prior Art】

25 A polymer electrolyte hydrogen-oxygen fuel cell is excellent in its output characteristics, whereby its application to e.g. automobiles, is expected. For

practical application of the above fuel cell, it is required to develop a fuel cell which provides a high output density and a high energy efficiency constantly over a long period of time even under such an operational
5 condition that the utilization ratios of a fuel and air are high.

The polymer electrolyte fuel cell usually has a construction such that a pair of power generation electrodes (a fuel electrode and an air electrode) face
10 each other via a polymer electrolyte, and the pair of electrodes and the electrolyte are bonded to form a membrane electrode assembly. A plurality of such assemblies are laminated via separators, and the entire assembly is clamped to be integral (stacked).

15 Here, the separator has channels for supplying a fuel gas and an oxidant gas (such as air) to the electrodes, and it serves as a partition plate between the adjacent two units. Accordingly, the separator is required to have characteristics such that the gas
20 permeability is low, it is light in weight, it is excellent in corrosion resistance and oxidation resistance when exposed to a steam atmosphere at a temperature in the vicinity of 150°C as the operational temperature of the fuel cell, it has good electrical
25 conductivity for a long period of time, and it can be mechanically processable. Further, the separator is required to be a good conductor for electricity and heat

in order to efficiently remove electricity and heat generated by a reaction of the cell out of the cell system.

As conventional separator materials, carbon type
5 bulk materials such as artificial graphite and glassy carbon, are known. However, the carbon type materials are poor in toughness and brittle, whereby the following problems are likely to occur when it is used as a separator under such a condition that a stress other than
10 a compression stress, or a mechanical shock, is likely to be exerted. Namely, the problems are such that the separator itself is broken, whereby the shape can not be maintained, cracking is likely to form, whereby air tightness can not be maintained, mechanical forming or
15 processing is difficult, whereby the processing cost tends to be high, and recycling is difficult.

As a means to solve the above problems, it has been proposed to use as a separator a molded product obtained by subjecting flat graphite powder particles so called
20 expanded graphite (such as Grafoil, tradename, manufactured by UCAR Co.) to dispersion treatment by e.g. an acid and adding a binder, followed by molding. This molded product is a flexible material which can be mechanically processable by e.g. pressing, whereby the
25 problems relating to the toughness and the mechanical shock resistance, are overcome. However, such a molded product has a problem such that the mechanical strength

is low, and the shape can hardly be maintained when made thin, or it is susceptible to deforming even when a small stress is applied thereto.

Further, the fuel cell is required to be small in size and light in weight and to provide a high output especially when it is used as a power source for an automobile, and it is necessary to increase a power per unit volume and to cool a heat generated by conducting electricity and by a reaction of the cell with a compact structure. Especially when a fluorine-containing polymer electrolyte having a high electrical conductivity, is used, this cooling will be essential, since the heat resistant temperature of the electrolyte is usually not so high. Cooling can be accomplished with the most compact structure when a fluid is permitted to flow at a high flow rate through a narrow and long channel formed in a separator, specifically, when water is permitted to flow under a high pressure. However, when the above-mentioned expanded graphite is used as the separator material, if the separator is maintained in the presence of water at a high temperature, the water is likely to penetrate into laminated particles of graphite, whereby the shape can not be maintained, thus leading to a problem that a long term reliability can not be attained.

As a means to solve the above-mentioned problems of the separator made of a carbon material, an attempt has been made to use a metal such as a surface-treated

stainless steel, titanium or aluminum as the separator material (e.g. EP0780916). When a metal is employed, the mechanical processing will be easy, the strength will be high even thin, the toughness, the mechanical shock resistance, the fluid shielding property, and thermal and electrical conductivity will be excellent. However, the metal material has problems such that the specific gravity is large (for example, 8.0 with stainless steel, 4.5 with titanium and 2.7 with aluminum), whereby the output per unit weight of the fuel cell tends to be low.

【Problems that the Invention is to Solve】

Therefore, the present invention has an object to provide a polymer electrolyte fuel cell having a separator which is made of a material which can easily be formed and processed, whereby the shape and air tightness can be maintained even when a stress other than a compression stress, a mechanical shock, vibration, etc., are exerted, and the initial good electrical conductivity can be maintained over a long period of time even when exposed to a steam atmosphere from room temperature to the vicinity of 150°C as the operational temperature of the fuel cell, and which is light in weight and industrially practical.

【Means of Solving the Problems】

The present invention provides a polymer electrolyte fuel cell comprising a plurality of membrane electrode assemblies laminated via separators, each assembly

comprising a membrane-form polymer electrolyte and a pair of a fuel electrode and an air electrode facing each other via the electrolyte, wherein the separator has a fuel gas channel for supplying a fuel gas to the fuel
5 electrode, an oxidant channel for supplying an oxidant to the air electrode and a fluid channel for removing a heat generated by a reaction out of the cell system, and the separator is made of a metal/non-metal composite material which has faces made of non-metal which are in contact
10 with the membrane electrode assemblies and side walls of the fluid channel which are made of metal.

In the present invention, the separator has a role to provide a fuel gas and an oxidant to the fuel electrode and the air electrode, respectively, and a role
15 to circulate a fluid for cooling to remove a heat generated by conduction of electricity and by a reaction of the cell out of the cell system. Here, air is used mainly as the oxidant, and accordingly, the electrode to which the oxidant is supplied, will be referred to as an
20 air electrode in this specification. As the fluid for cooling, water is preferred, since the heat can thereby be efficiently removed. Further, the separator has a role to shield a gas or moisture to prevent permeation of the gas or moisture between the adjacent two membrane
25 electrode assemblies and a role to transmit the generated electric current.

The fuel gas channel and the oxidant channel usually

consist of grooves formed by ribs formed on the surface of the separator. Such a gas channel may consist of a plurality of linear or curved grooves, but a meandering groove may be formed over the entire surface of the separator so that it has only one inlet and one outlet for a gas.

In the present invention, the separator is made of a metal/non-metal composite material, and at least side walls of the fluid channel for cooling are made of metal. Accordingly, even when the above side walls are exposed to water as the fluid for cooling at a temperature of up to about 150°C as the operational temperature of the fuel cell, the water for cooling will not penetrate into the interior of the separator, and the shape of the separator can be maintained.

Further, in the separator in the present invention, at least faces which are in contact with the membrane electrode assemblies, are made of non-metal. The separator has a role to conduct an electric current, the above non-metal is preferably made of a highly electrically conductive carbon material. Particularly preferably, it is made of a molded product obtained by subjecting expanded graphite particles to dispersion treatment with e.g. an acid, adding a binder, followed by molding. Such a molded product is readily mechanically processable by e.g. pressing and is light in weight.

In order to reduce the weight of the separator in

the present invention, it is preferred that the proportion of the non-metal in the metal/non-metal composite material is as large as possible. Accordingly, from this viewpoint, it is preferred that all other than
5 the side walls of the fluid channel is composed of the non-metal. However, from the production efficiency of the separator and high strength of the separator, it is preferred that a layer made of metal having a fluid channel internally (hereinafter referred to as a metal
10 layer) is sandwiched between a pair of layers made of non-metal having gas channels on their surfaces (hereinafter referred to as non-metal layers) and bonded to constitute the separator. Here, the pair of non-metal layers sandwich the metal layer so that the gas channels
15 will be located on the surfaces.

In the present invention, the metal constituting the separator is preferably one member selected from the group consisting of a metal containing aluminum in an amount of at least 80 wt%, a metal containing titanium in
20 an amount of at least 80 wt% and stainless steel. Here, the metal includes an alloy. These metals have high strength even when they are thin, and they are excellent in reliability against a dynamic shock such as vibration or mechanical shock and against a static mechanical load
25 such as tension or compression. Further, they are capable of maintaining their shapes even when exposed to a fluid such as water at a high temperature for a long

period of time, and they are excellent also in thermal and electrical conductivity.

In the above-mentioned metal containing aluminum or the metal containing titanium, if the content of aluminum or titanium as the main component, is less than 80 wt%, the specific gravity of metal constituting the separator tends to be large, such being undesirable. The content of aluminum or titanium is particularly preferably from 90 to 98 wt%.

10 The metal containing aluminum as the main component is light in weight, easy for recycling and mechanically readily processable, such being desirable. As the aluminum alloy, an alloy of aluminum with at least one metal selected from the group consisting of magnesium, 15 manganese, silicon, copper, nickel, lithium, zinc, lead, bismuth, titanium and tin, is preferred. For example, duralumin, silumin, hydrosodium or anticorrosional may be mentioned.

20 Further, the metal containing titanium as the main component has high mechanical strength per unit weight and relatively high corrosion resistance, such being desirable. For example, an alloy of titanium with at least one metal selected from the group consisting of aluminum, iron, vanadium, molybdenum, manganese, chromium, 25 zirconium, tin, silicon, palladium and tantalum, is a corrosion resistant alloy and preferred.

Further, stainless steel has high mechanical

strength per unit volume and relatively high corrosion resistance, such being desirable. The stainless steel is not particularly limited, and any one of austenite type, ferrite type and martensite type, may be employed. From
5 the viewpoint of corrosion resistance, austenite type stainless steel is particularly preferred.

It is preferred that a coating film containing ceramics and having a resistivity of at most $3 \times 10^{-4} \Omega \cdot \text{cm}$, particularly from 3×10^{-6} to $1 \times 10^{-4} \Omega \cdot \text{cm}$, is formed on the
10 surface of side walls of the fluid channel made of the metal, i.e. on the surface of the above metal which is in contact with the fluid. By the formation of the coating film containing ceramics, deterioration of the metal surface by oxidation can be prevented. If the
15 resistivity of the coating film exceeds $3 \times 10^{-4} \Omega \cdot \text{cm}$, the resistivity of the overall fuel cell tends to be high, whereby it may happen that the energy can not efficiently be taken out. This coating film may be a coating film composed solely of ceramics, or a coating film having
20 ceramics dispersed in metal.

Further, it is preferred that a layer containing ceramics and having a resistivity of at most $3 \times 10^{-4} \Omega \cdot \text{cm}$, is disposed at the interface between the metal and the non-metal constituting the separator. In the polymer
25 electrolyte fuel cell, water is formed at the air electrode along with the reaction of the cell. Further, usually, in order to maintain the ion conductivity of the

polymer electrolyte, the fuel gas and the oxidant gas are supplied to the respective channels, as wetted so as not to dry the electrolyte. Such formed water and the steam contained in the gas will not flow rapidly and in a large
5 amount like water as the fluid for cooling, whereby even if the non metal portion of the separator is exposed to them, the shape of the separator will not be disintegrated.

However, such water contents are likely to penetrate
10 through the non metal portion of the separator and reach the interface between the metal and the non-metal. Consequently, it is likely that the surface of the metal in contact with the non-metal will be oxidized, whereby the electrical conductivity tends to decrease. Here, if
15 a layer made of ceramics excellent in the durability as compared with the metal, is present at the interface between the metal and the non-metal, deterioration of the metal can be prevented. Further, if the resistivity of this layer made of ceramics exceeds $3 \times 10^{-4} \Omega \cdot \text{cm}$, the
20 resistivity of the overall fuel cell tends to be high, whereby the energy may not efficiently be taken out. The resistivity of this layer is particularly preferably from 3×10^{-6} to $1 \times 10^{-4} \Omega \cdot \text{cm}$. This layer may be a layer composed solely of ceramics or a layer having ceramics dispersed
25 in metal.

【Mode of Carrying out the Invention】

Now, practical embodiments of the present invention

will be described with reference to Figs. 1 and 2.

Fig. 1(a) is a plan view of a separator 7 as viewed from the air electrode side or from the fuel electrode side, and Fig. 1(b) is a cross-sectional view of the separator 7 taken along the line A-A' of Fig. 1(a). In the separator 7, a metal layer 2 is disposed between a non-metal layer 1a and a non-metal layer 1b.

On the surfaces of the non-metal layers 1a and 1b, gas channels 3a and 3b are formed, respectively. The gas channels 3a and 3b are composed of grooves formed on the surfaces of the non-metal layers 1a and 1b as shown in Fig. 1(b). The grooves have one portion each for the inlet and the outlet of the gas, and they are formed as meandering over the entire surfaces of the non-metal layers 1a and 1b, respectively. The faces having the grooves are in contact with the membrane electrode assemblies, and gases are supplied to the membrane electrode assemblies through the gas channels 3a and 3b.

On the surfaces of the non-metal layers 1a and 1b, the portions which are in contact with the membrane electrode assemblies, are preferably surface-treated with a water repellent containing fluorine. By the presence of the water repellent on the surfaces, it is possible to prevent retention of water formed by the reaction at the membrane electrode assemblies at the non-metal layers 1a and 1b, whereby supply of the fuel gas or the oxidant gas to the membrane electrode assemblies can be maintained

properly. Further, it is possible to prevent swelling of the non-metal layer due to penetration of water into the interior of the non-metal layer.

In the metal layer 2, a fluid channel 4 for cooling is internally formed, and this metal layer 2 will be described in further detail with reference to Figs. 2(a) and (b). Fig. 2(a) is a plan view of a metal substrate 2a constituting the metal layer 2 of the separator, and Fig. 2(b) is a cross-sectional view along line B-B' of Fig. 2(a) and a cross-sectional view of a metal substrate 2b paired with the metal substrate 2a. In the metal substrate 2b, a groove which constitutes a fluid channel for cooling, is formed symmetrically to the metal substrate 2a.

The metal layer 2 is obtained by putting together the metal substrate 2a and the metal substrate 2b to be paired, so that the faces on which grooves are formed, face to each other so that the positions of grooves 4a and 4b to constitute the flow channel for cooling will agree, and bonding contacting faces 7a and 7b. Here, as the bonding method, welding, soldering, diffusion bonding or bonding with an adhesive may, for example, be mentioned.

Coating films 5a and 5b containing ceramics are formed on the surfaces of the grooves 4a and 4b constituting the fluid channel for cooling, respectively, formed on the metal substrates 2a and 2b. Accordingly,

the fluid for cooling will be in direct contact with such coating films 5a and 5b.

Further, layers 6a and 6b which contain ceramics and which are disposed at the interfaces between the metal
5 layer and the non-metal layer, are formed on the surfaces of the sides of the metal substrates 2a and 2b on which the fluid channels 4a and 4b for cooling are not formed.

The coating films 5a and 5b and the layers 6a and 6b disposed at the interfaces between the metal layer and
10 the non-metal layer, are layers each containing ceramics and having a resistivity of at most $3 \times 10^{-4} \Omega \cdot \text{cm}$, and their constituting materials may be the same or different. Preferred components and method for forming them are common and will be described in the following summary as
15 a description of the coating film.

The ceramics contained in the coating film is preferably one which is excellent in corrosion resistance even in a steam atmosphere within a temperature range of from room temperature to the vicinity of 150°C in which
20 the fuel cell is operated and which is capable of maintaining a stabilized resistivity, and a nitride, a boride, a carbide, a silicide or a phosphide of a metal, or a composite compound thereof, is preferred. As such a metal, a rare earth element such a yttrium, lanthanum,
25 cerium or samarium, iron, nickel, cobalt, chromium, niobium, tantalum, titanium, zirconium, vanadium, hafnium, palladium, tungsten or molybdenum, can preferably be

employed. As the ceramics, specifically, titanium carbide, titanium boride, titanium nitride, tungsten silicide, or tantalum nitride, is, for example, preferred. These may be used alone or may be used in an admixture of
5 two or more of them.

In the case of the coating film having ceramics dispersed in metal, such metal is preferably one excellent in corrosion resistance, and it is preferably at least one member selected from the group consisting of
10 a platinum group element, gold, nickel and tungsten. The content of ceramics in this coating film is preferably from 1 to 90 wt%, particularly preferably from 10 to 60 wt%.

The thickness of the coating film to be formed on
15 the metal surface is preferably from 0.1 μm to 0.3 mm, particularly preferably from 1 μm to 0.1 mm. If it is thinner than 0.1 μm , it tends to be difficult to form a continuous coating film or layer having adequate corrosion resistance. If it is thicker than 0.3 mm, even
20 if a material having a low resistivity is employed, the resistance of the coating film or layer tends to be high, whereby it tends to be difficult to take out the energy efficiently from the fuel cell. Further, the output per unit volume of the fuel cell tends to be small.

25 The coating film to be formed on the metal surface, may be formed in a pattern such as a strip pattern, a line pattern, an island pattern or a dotted pattern. The

coating film or layer may be formed regularly or irregularly, and the thickness of the coating film may be uniform or non-uniform.

As a method for forming the coating film, a thick
5 film-forming method such as a screen printing method, a doctor blade method, a spray method, a flame spraying method or an ion plating method, or a thin film-forming method such as a CVD method or a PVD method such as a sputtering method, may be employed. Otherwise, the
10 coating film having ceramics dispersed in metal may be formed by a dispersion plating method wherein plating is carried out by using ceramics as a dispersant and a metal as a binder. The above-mentioned dispersion plating method is simple and preferred in a case where the
15 production is carried out on an industrial scale, since the coating film can firmly be bonded to the metal substrate without peeling of the coating film even when the thermal expansion coefficient is substantially different between the ceramics and the metal for
20 constituting the separator.

Further, an interlayer may be present between the above-mentioned coating film and the metal substrate in order to improve the adhesiveness. As such an interlayer, a layer of a metal excellent in corrosion resistance such
25 as titanium, nickel, cobalt, gold or platinum, may, for example, be mentioned.

The thickness of the metal layer 2 is required to be

at least a thickness whereby the non-metal layers 1a and 1b can sufficiently be shielded from the fluid for cooling. Further, in order to increase the energy density per unit weight or unit volume of the fuel cell, the thinner the thickness of the metal layer 2, the better. Accordingly, the thickness of the metal layer 2 is preferably from 1 μ m to 2 mm, particularly preferably from 10 μ m to 1 mm.

As a method for bonding the non-metal layers 1a and 1b and the metal layer 2, it is preferred to employ, for example, bonding with silver solder.

The membrane electrode assembly is obtained, for example, as follows. A mixture of a catalyst having a metal catalyst such as platinum supported on a carbon material such as activated carbon, with a water repellent, etc., is coated or sprayed on a carbon paper to prepare a gas diffusion electrode. Two sheets of such an electrode are prepared as a fuel electrode and an air electrode. Then, the fuel electrode and the air electrode are disposed to face each other, and a polymer electrolyte made of an ion exchange membrane is interposed therebetween, followed by bonding by a hot pressing method to unify the air electrode/the polymer electrolyte/the fuel electrode to obtain a membrane electrode assembly.

The above ion exchange membrane is preferably made of a fluorine-containing carbon polymer having sulfonic

groups, from the viewpoint of the chemical and electrical stability.

When the polymer electrolyte fuel cell of the present invention is used, for example, in an environment
5 where vibration or shock is likely to be imparted in the operation as in an application to an engine for an automobile, the long term reliability of the operation of the cell is remarkably improved as compared with a fuel cell employing a separator made of a carbon material.

10 Further, by forming a layer containing ceramics at a portion of the separator which is in contact with the fluid for cooling or at the interface between the non-metal layer and the metal portion, a constant performance of the fuel cell not different from the initial stage,
15 can be maintained over a long period of time.

【Examples】

Now, the present invention will be described in further detail with reference to Examples (Examples 1 to 3) and Comparative Examples (Examples 4 and 5). However,
20 the present invention is by no means restricted thereto.

EXAMPLE 1

A metal substrate made of an aluminum alloy (aluminum content: 96 wt%) of alloy No. A5052 as stipulated by JIS-H4000 and having a width of 150 mm, a
25 height of 150 mm and a thickness of 0.3 mm, was subjected to pressing to form a groove for a fluid channel for cooling with a width of 1.5 mm and a depth of 0.15 mm in

a pattern as shown in Fig. 2(a). On the surface of this groove and on the surface of the side where no groove was formed, a Ti layer having a thickness of 0.5 μm was formed as a primer layer by an ion plating method, and a
5 titanium nitride coating film having a thickness of 10 μm was further formed thereon. The resistivity of this titanium nitride coating film was $2 \times 10^{-4} \Omega \cdot \text{cm}$.

The same metal substrate as the above metal substrate except that the groove is formed symmetrically,
10 was prepared. As shown in Fig. 2(b), these two sheets of metal substrates were overlaid each other so that the sides on which the grooves were formed, faced each other so that the positions of the grooves of these two sheets of metal substrates agreed to each other, and bonded by
15 welding to obtain a metal layer to constitute a separator.

Then, using an expanded graphite having a thickness of 1.5 mm (tradename: Grafoil, manufactured by UCAR Co.), a groove to constitute a gas channel with a width of 2 mm and a depth of 0.35 mm, was formed on one side by
20 pressing in a pattern as shown in Figs. 1(a) and (b), to obtain a non-metal layer. Here, as shown in Fig. 1(a), the groove was formed as meandering, as shown in Fig. 1(a), whereby the distance between the adjacent grooves formed in parallel by meandering, was set to be 3 mm.
25 Two sheets of such a non-metal layer were prepared so that the positions of the grooves were different, as shown in Fig. 1(b).

The above two non-metal layers were put together so that the faces on which no grooves were formed, faced each other, with the above metal layer interposed therebetween. In this state, three layers of non-metal layer/metal layer/non-metal layer were press-bonded to obtain a separator.

Then, on one side of a carbon cloth, a catalyst having platinum supported on carbon black was coated to form a catalyst layer thereby to obtain a gas diffusion electrode. Two sheets of such a gas diffusion electrode were disposed so that the faces on which the catalyst layers were formed, faced each other, and a perfluorocarbon sulfonic acid type ion exchange membrane (Flemion R, tradename, manufactured by Asahi Glass Company, Limited) was sandwiched between them, followed by hot pressing to obtain a membrane electrode assembly.

20 sheets of the above separator and 19 sheets of the above membrane electrode assembly were alternately laminated, and the entirety was clamped to obtain a polymer electrolyte fuel cell. Ten such fuel cells were prepared, and with respect to each of them, a dropping test was repeated ten times. The dropping test was carried out in such a manner that with the side of separator facing downwardly, it was dropped straightly from a position at a height of 50 cm onto a steel plate having a thickness of 5 cm. After the dropping test, air was permitted to flow through the gas channel of the

separator under a pressure of 5 kg/cm^2 to carry out a gas leak test, whereby with 10 samples, there was no problem with respect to the air tightness.

One of the above fuel cells was taken out, and as a
5 fuel gas, hydrogen gas under normal pressure and as an oxidant gas, air under normal pressure, were permitted to flow, so that the utilization ratios would be 70% and 40%, respectively. Further, tap water was permitted to flow through the fluid channel for cooling under a water
10 pressure of 5 kg/cm^2 for cooling, and the fuel cell was operated at 80°C at a current of 140 A, whereby the initial voltage was measured as the initial output. Then, the current was continuously conducted, and the voltage after expiration of 5000 hours was measured, and the
15 ratio (%) to the initial voltage value was calculated as the relative output. The results are shown in Table 1.

EXAMPLE 2

Using a metal substrate made of titanium having a purity of 99.4% and having a width of 150 mm, a height of
20 150 mm and a thickness of 0.2 mm, a groove to constitute a fluid channel for cooling was formed on one side by etching in the same pattern as in Example 1. On the surface of this groove and on the surface of the side on which no groove was formed, plating was applied by means
25 of a nickel watt bath having particles of titanium nitride dispersed (the distribution range of the crystal particle sizes: 0.1 to 3 μm , resistivity: $4.0 \times 10^{-5} \Omega \cdot \text{cm}$)

to form a coating film having a thickness of 10 μm . The resistivity of this titanium nitride/nickel coating film was $2.0 \times 10^{-5} \Omega \cdot \text{cm}$.

The same metal substrate as the above metal
5 substrate made of titanium except that the groove is formed symmetrically, was prepared, and in the same manner as in Example 1, the pair of metal substrates were bonded to obtain a metal layer to constitute a separator.

A polymer electrolyte fuel cell was prepared in the
10 same manner as in Example 1 except that the above metal layer was employed, and the same dropping tests as in Example 1 were carried out, and further the gas leak test was carried out, whereby there was no problem with respect to the air tightness. Then, durability test was
15 carried out in the same manner as in Example 1 to calculate the relative output after expiration of 5,000 hours. The results are shown in Table 1.

EXAMPLE 3

Using a metal substrate made of SUS316 and having a
20 width of 150 mm, a height of 150 mm and a thickness of 0.15 mm, a groove to constitute a fluid channel for cooling was formed on one side in the same manner as in Example 1. On the surface of this groove and on the surface of the side on which no groove was formed, a
25 coating film having a thickness of 0.3 μm made of molybdenum silicide, was formed by sputtering. The resistivity of this molybdenum silicide coating film was

$2.2 \times 10^{-5} \Omega \cdot \text{cm}.$

The same metal substrate to be paired as the above metal substrate made of SUS316 except that the groove was formed symmetrically, was prepared, and in the same manner as in Example 1, the pair of metal substrates were bonded to obtain a metal layer to constitute a separator.

A polymer electrolyte fuel cell was prepared in the same manner as in Example 1 except that the above metal layer was employed, the same dropping tests as in Example 1 were carried out, and further the gas leak test was carried out, whereby there was no problem with respect to the air tightness. Then, durability test was carried out in the same manner as in Example 1 to calculate the relative output after expiration of 5,000 hours. The results are shown in Table 1.

EXAMPLE 4

Instead of the substrate made of an aluminum alloy in Example 1, a substrate made of expanded graphite having the same size as said substrate, was employed, and a groove to constitute a fluid channel for cooling was formed on one side in the same pattern as in Example 1.

The same substrate to be paired as the above substrate made of expanded graphite except that the groove was formed asymmetrically, was prepared, and the pair of substrates were put together so that the faces of the pair of substrates on which the grooves were formed, faced each other so that the positions of the grooves

agreed, and a polymer electrolyte fuel cell was prepared in the same manner as in Example 1 except that this assembly was used instead of the metal layer. The same dropping test as in Example 1 was carried out, and

5 further the gas leak test was carried out, whereby there was no problem with respect to the air tightness. Then, durability test was carried out in the same manner as in Example 1 to calculate the relative output after expiration of 5,000 hours. The results are shown in

10 Table 1.

EXAMPLE 5

A layer having the same size and the same shape as the non-metal layer in Example 1 was prepared by using the aluminum alloy used in Example 1. A separator was
15 prepared in the same manner as in Example 1 except that this layer was used instead of the non-metal layer, and the bonding with the metal layer was carried out by mechanical processing.

A polymer electrolyte fuel cell was prepared in the
20 same manner as in Example 1 except that this separator was employed. The same dropping test as in Example 1 was carried out, and further, the gas leak test was carried out, whereby there was no problem with respect to the air tightness. Then, durability test was carried out in the
25 same manner as in Example 1 to calculate the relative output after expiration of 5,000 hours. The results are shown in Table 1.

【Table 1】

	Relative output (%)
Example 1	98
Example 2	96
Example 3	97
Example 4	60
Example 5	85

【Effects of the Invention】

According to the present invention, even when the
5 polymer electrolyte fuel cell is used under an
environmental condition where a stress other than the
compression stress or a mechanical shock is imparted, the
separator will not be destroyed, cracking will not form,
and the air tightness of the fuel cell can be maintained
10 while maintaining the initial shape. Further, the
separator in the present invention is easy for mechanical
shaping and processing and can readily be recycled.
Further, the separator in the present invention is light
in weight as compared with a conventional separator made
15 of metal. Accordingly, the polymer electrolyte fuel cell
of the present invention is light in weight, provides a
high output and is excellent in durability for a long
period of time.

【Brief Explanation of the Drawings】

20 【Figure 1】

(a) is a plan view of a separator of an embodiment

of the present invention. (b) is a cross-sectional view taken along line A-A' of Fig. 1(a).

【Figure 2】

(a) is a plan view of a metal substrate to form the metal layer of the separator of an embodiment of the present invention. (b) is a cross-sectional view of the metal substrate taken along line B-B' of Fig. 2(a) and the cross-sectional view of a metal substrate paired with said metal substrate.

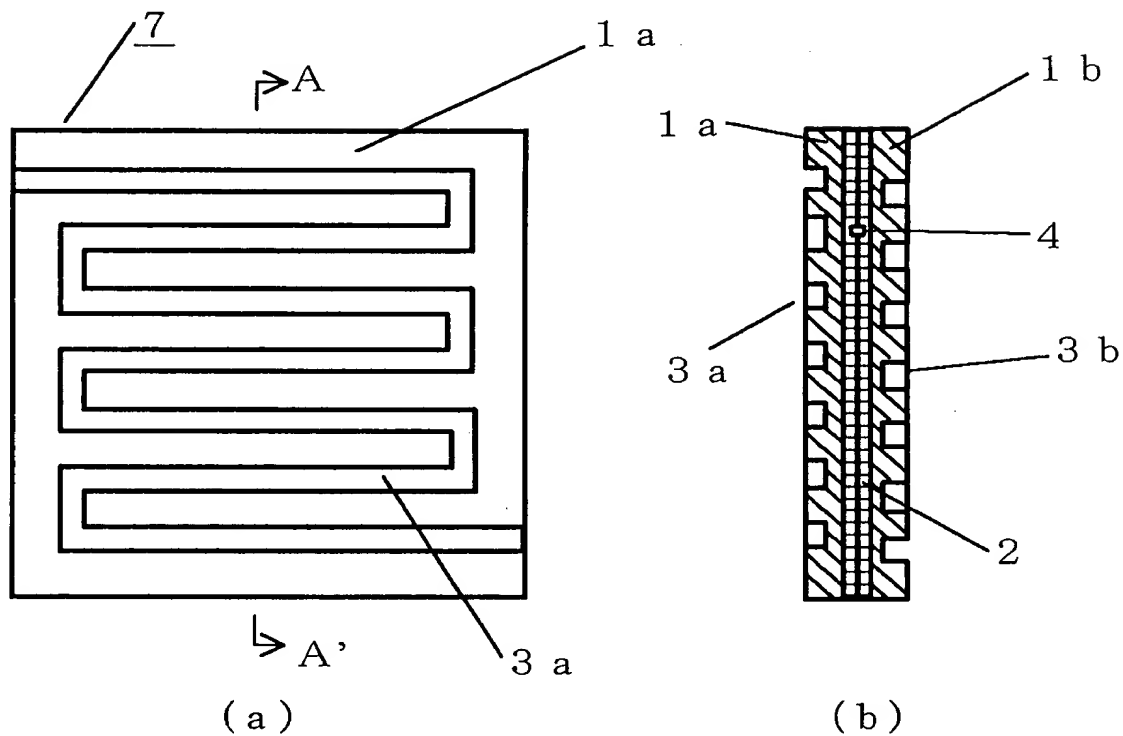
10 【Explanation of the Reference Numerals】

- 1a, 1b: Non-metal layers
- 2: Metal layer
- 2a, 2b: Metal substrates
- 3a, 3b: Gas channels
- 15 4: Fluid channel for cooling
- 4a, 4b: Grooves constituting a fluid channel for cooling
- 5a, 5b: Coating films
- 6a, 6b: Layers disposed at the interface between a metal layer and a non-metal layer
- 20 7: Separator

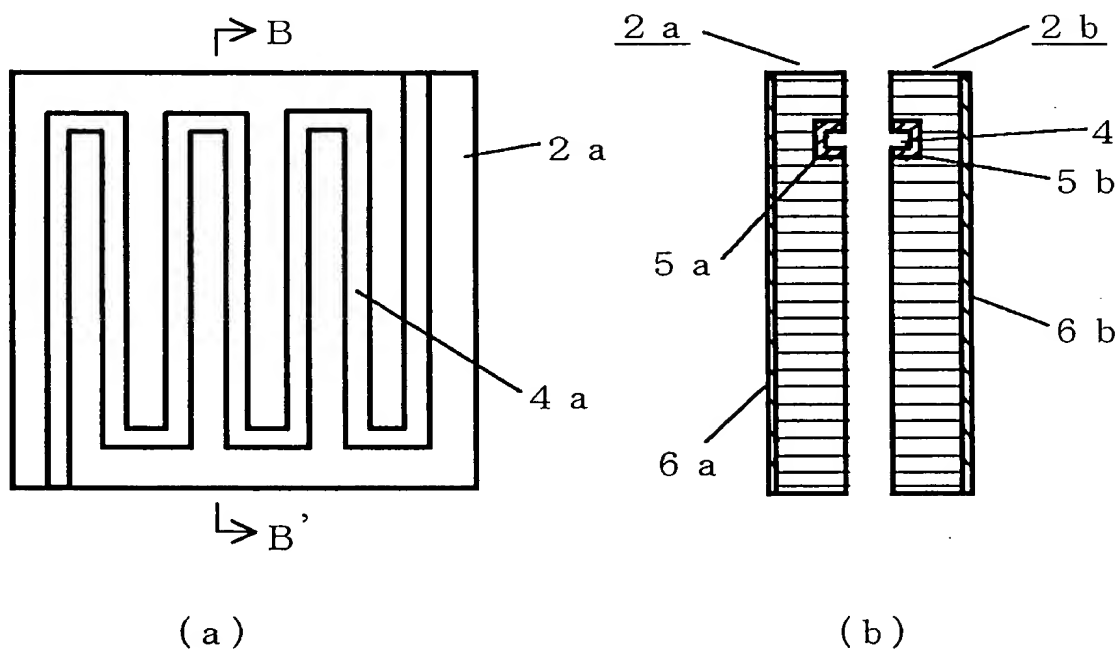


【TYPE OF DOCUMENT】 DRAWING

【Fig. 1】



【Fig. 2】



【TYPE OF DOCUMENT】

ABSTRACT

【SUMMARY】

【OBJECT】

It is an object to provide a polymer electrolyte
5 fuel cell which is excellent in the dimensional stability
and air-tightness and light in weight, and can maintain a
high performance constantly for a long time.

【MEANS OF SOLVING PROBLEMS】

A polymer electrolyte fuel cell using a separator
10 made of a metal/non-metal composite material, wherein at
least side walls of a fluid channel for cooling are made
of metal and at least faces which are in contact with
membrane electrode assemblies are made of non-metal.

【SELECTED FIGURE】

Fig. 1